LASER MODULATION AT THE ATOMIC LEVEL

Monthly Report No. 2

Date of this Report: 10 September 1964
Period covered: 1 August 1964 to 31 August 1964

Submitted to
National Aeronautics and Space Administration
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GENERAL DYNAMICS | ELECTRONICS

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LASER MODULATION AT THE ATOMIC LEVEL

Purpose

Research on methods of influencing internally the radiating centers of active laser materials in order to achieve laser modulation is the principal objective of the work carried out under this contract.

Summary

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The determination of gain in ruby as a function of magnetic field was continued, with some indication of the appreciable influence of excited state absorption upon the measurements. Planning for the initiation of Phase II suggests substitution of $YAG:Nd^{3+}$ for $CaF_2:Dy^{2+}$ as the initial laser material to be studied in that phase. Preparations for the experimental observation of the Zeeman splitting of the laser emission in a homogeneous pulsed field are described.

Man-Hours Worked

The total number of man-hours worked during the reporting period is 601.5 hours.

I. INTRODUCTION

During the month of August 1964 work continued on the determination of gain in ruby laser crystals as a function of inhomogeneous magnetic field intensity. Improvements in the experimental arrangement and problems encountered from apparently significant excited state absorption in pursuing this aspect of Phase I are presented in Part II of this report. Modifications in planning for Phase II required by advances in the field of rare earth doped laser materials and preparations for the study of Zeeman splitting in the YAG:Nd^{3+} laser transition are presented in Part III.

II. MEASUREMENTS ON RUBY

A. Ground State Absorption

As discussed in Monthly Progress Report No. 1, the bleaching of ground state absorption under optical pumping is to be used as a means for measuring the degree of population inversion at laser threshold as a function of magnetic field. Two high optical quality ruby rods have been obtained from the Linde Crystal Products Division of Union Carbide Corporation for these experiments. Rods having 0° and 90° orientation, 1-1/2 inches long by 1/4 inch OD, were cut from the same disc boule in order to make comparisons between the magnetic field effects on the two orientations as independent of material variations as possible. A 90° polished window 1.22 mm thick was obtained from a section of the boule adjacent to the 90° rod. This window has been used to obtain the ground state absorption spectra, and will also be used to measure the R_1 linewidth as a function of temperature in order to permit more accurate comparison between experiment and theory.

The absorption measurements have been made using a Perkin-Elmer 13U dual-beam spectrophotometer. Difficulties were first encountered in obtaining reproducible spectra because the spectrometer acts as a partial polarizer (the transmittance for the horizontally polarized component is about 20% greater than that for the vertically polarized component, due mainly to selective reflection from the prism faces). The procedure finally adopted utilized a combination of polarizer and analyser with the ruby window at the sample position between them. The polarization of the transmitted light in the parallel orientation of the polaroids was horizontal. The polaroids were first crossed, the ruby window inserted to obtain extinction (insuring $E \perp C$ to $\pm 0.5^\circ$), and the analyser was then set parallel to the polarizer for the measurement of the absorption spectrum with $E \perp C$. The ruby was then rotated 90° and adjusted for extinction to obtain the spectrum for $E \perp C$. The linear extinction coefficient α was then calculated from the relation

$$\alpha = \frac{1}{L}\left[\ln \frac{I}{I_o} - 2\ln(1 - r) - \delta\right],$$

where L is the sample thickness, I/I_o is the ratio of transmitted intensities, r is the reflectivity at normal incidence $[r = (1 - n)^2/(1 + n)^2]$, and δ is a correction term, assumed to be wavelength independent, due mainly to scattering from the surfaces. The correction term was first assumed to be zero, and then determined by the correction needed to make $\alpha = 0$ in the high transmission band between 6500 Å and 6900 Å. For various measurements δ proved to be between 2% and 3%, which is reasonable for scattering from two polished surfaces. The reflectivity was calculated from the Cauchy equations for sapphire derived from values of the index of refraction reported in the A. I. P. Handbook.* These equations are:

$$n_{\perp} = 1.7513 + 5.803/\lambda^2,$$

$$n_{\parallel} = 1.7438 + 5.580/\lambda^2,$$

where λ is the wavelength in microns. The spectra for $\alpha_{\perp}$ and $\alpha_{\parallel}$ obtained from a number of runs are shown in Figs. 1 and 2, respectively. The spectral range of these measurements was limited by the range over which the polaroids had a reasonably high rejection ratio.

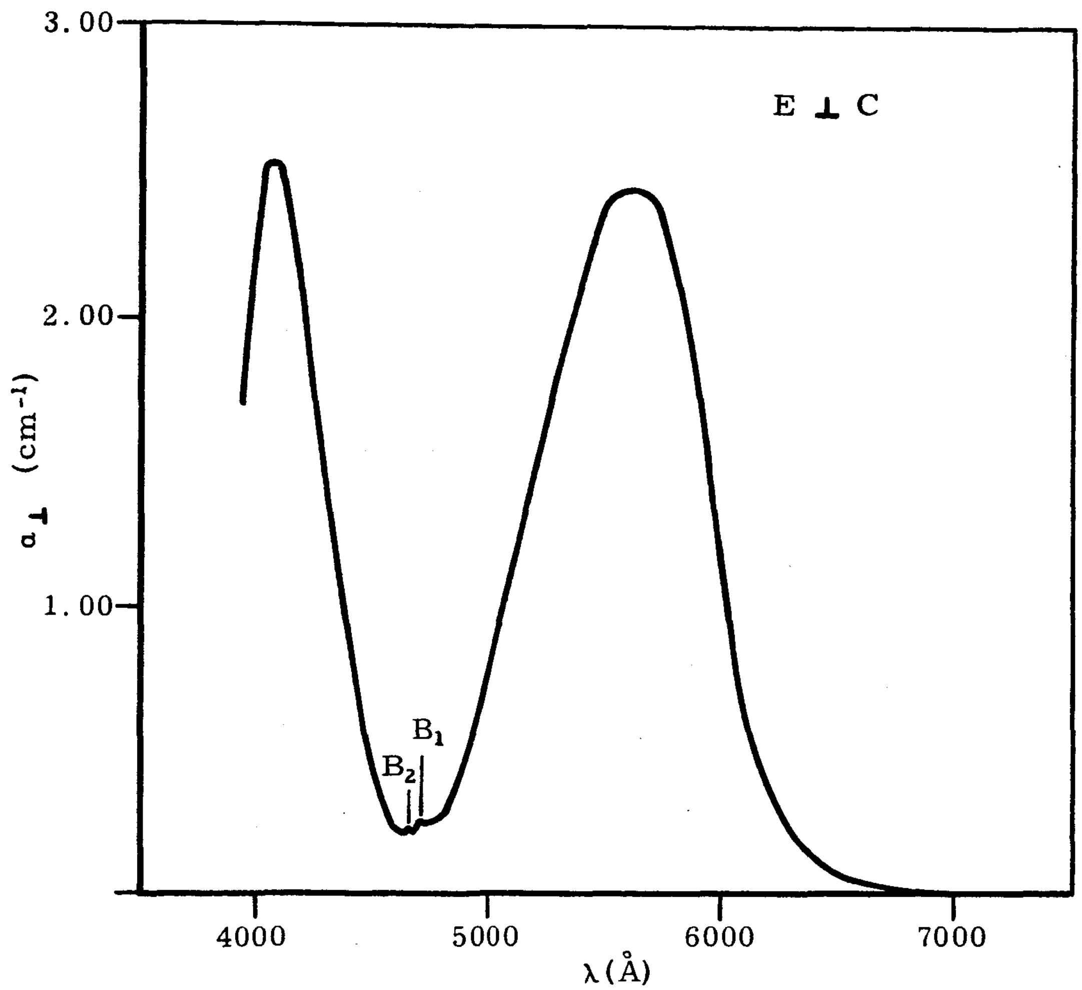

Fig. 1. Ground state linear absorption spectrum, electric vector perpendicular to optic axis (ordinary ray).

*American Institute of Physics Handbook (McGraw-Hill, New York, 1957).

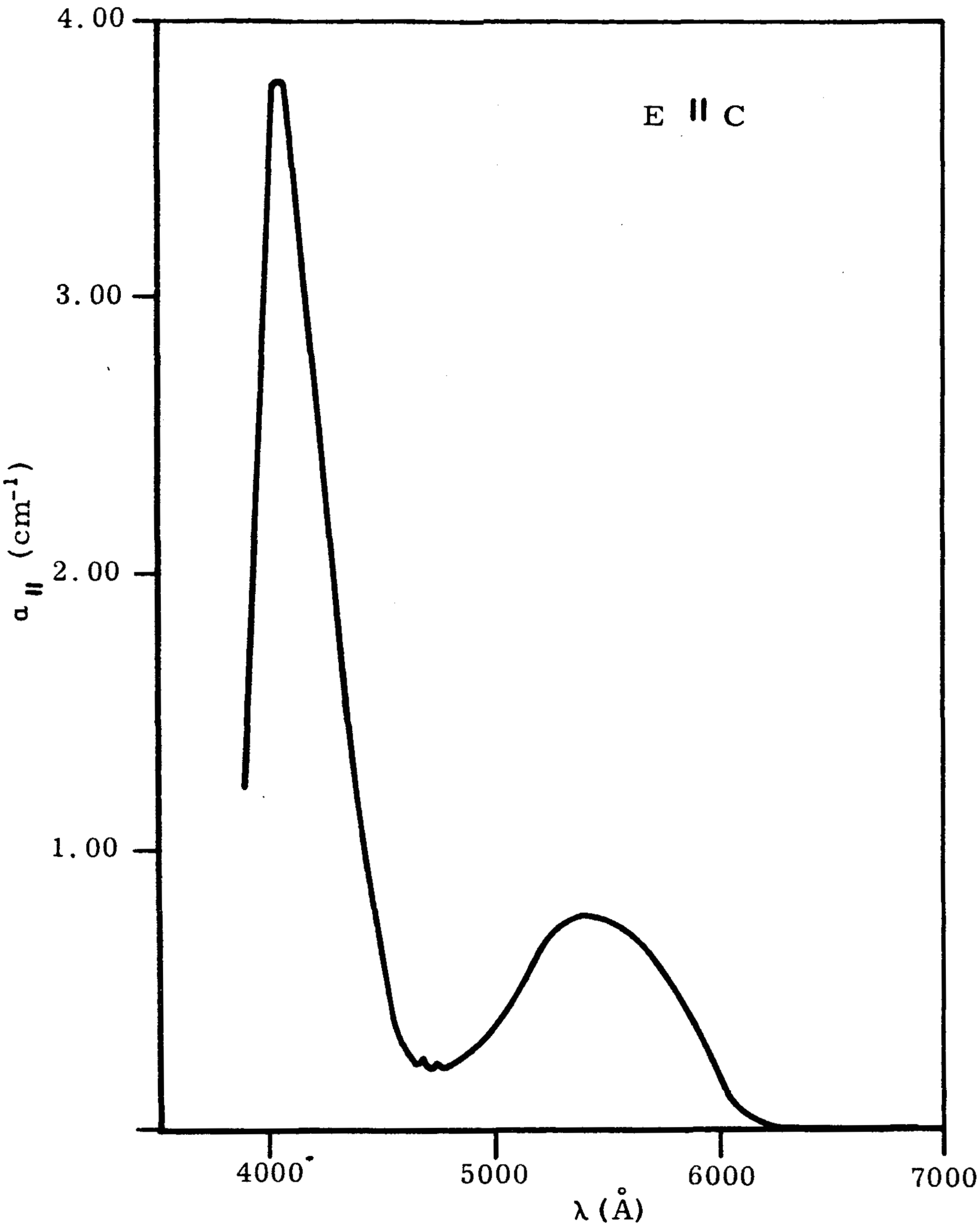

Fig. 2. Ground state linear absorption spectrum, electric field parallel to optic axis (extraordinary ray).

The values of peak absorption in the 5500 Å region were compared with the absolute absorption cross-section data of Dodd, Wood and Barnes.* The Cr^{3+} ion concentration was found to be 0.050±0.002 wt%, the concentration specified to Linde, and the agreement may be taken as an indication of the correctness of the absorption spectra which we have obtained.

*D. M. Dodd, D. L. Wood, and R. L. Barnes, J. Appl. Phys., 35, 1183 (1964).

B. 0° Rod: Threshold and Near Field Intensity

The 0° oriented laser rod was mounted on the evacuated holder and tested for laser threshold and near field emission pattern in the temperature range where we plan to make the magnetic field measurements. Threshold was measured by determining integrated laser emission energy as a function of pump energy and extrapolating the resulting linear relationship to zero laser energy. Table I gives the results for four ruby temperatures.

TABLE I

Threshold for 0° ruby rod, one end multilayer dielectric coated, other end uncoated

Temperature (°K)	118	122	131	138
Pump lamp input energy in Joules	602	605	631	660

The dielectric coatings supplied by Linde proved to have very poor adhesion and had to be stripped off the rods. They are being replaced by aluminum coatings, which can be deposited in our laboratory.

The near field emission patterns were photographed at 4X magnification of the front ruby face, using Kodak infrared sensitive sheet film. At 1.05 times threshold, the laser emission consisted of a fairly symmetrical mode pattern centered on the rod axis and having a radius of about 1.5 mm for the major intensity peaks. At 1.15 times threshold, the region of oscillation had extended to about 2.5 mm, with a less clearly defined mode pattern. These measurements indicate that a probe beam diameter less than 1.5 mm passing through the central axis of the ruby should encounter a fairly uniform radial pump flux distribution. The actual probe beam has a maximum diameter of less than 1 mm within the ruby rod.

C. Transmission Apparatus Modification

Tests of the pulsed light transmission apparatus showed that serious random errors were being introduced by fluctuations in the arc position within the end window flashlamp which was being used as the light source. The probe light, therefore has been changed to a PEK X-80, high pressure Xenon compact arc lamp. The lamp is operated at reduced dc current and is pulsed by an ignitron-switched 0.05 μf capacitor charged to 10 kv. The output light flash has a rise time of about 5 μsec and a fall time of about 20 μsec, providing adequate time resolution for our measurements. The smaller source diameter of the arc lamp permits better beam collimation than was possible with the flashlamp, and the continuous light output greatly simplifies accurate alignment of the optical system.

The light pipe has been removed from the monitor photomultiplier so that it now monitors the entire beam which passes through the ruby, rather than just a portion of that beam. This has considerably improved the reproducibility of the I/I_o intensity ratio measurements.

D. Preliminary Transmission Measurements

The first experiments on measurement of bleaching have yielded results which force us to abandon the assumption of negligible excited state absorption, which had been made in the discussion of the theory of these experiments presented in the first monthly report. Measurements made at the end of the present reporting period indicate appreciable excited state absorption at room temperature in the region from 6800 Å to 5000 Å in which we were planning to make our measurements. Fortunately, the excited state absorption appears to have a very slow variation with wavelength, so that it should be possible to determine the correction required and proceed with the measurements. Measurements of Gires and Mayer* tend to agree with our results, although their data do not extend much above 5000 Å, the region of interest to us. The experiments of Brand et al.** seem to imply that the excited state absorption in the vicinity of 6943 Å is much greater at low temperatures than at room temperature, and this could certainly have important consequences for our measurements. Since our measurements are incomplete, further discussion of these questions will be postponed to the next monthly report.

*F. Gires and G. Mayer, Quantum Electronics, Proceedings of 3rd International Conference, 841 ff, Columbia University Press, New York, 1964.

**F. A. Brand, H. Jacobs, S. Weitz, and C. LoCascio, Proc. IEEE, 52, 417 (1964).

III. PREPARATIONS FOR PHASE II

A. Current Status of Narrow Line Laser Materials

At the time that the proposal for the present contract was written $CaF_2:Dy^{2+}$ appeared to be a very favorable candidate for the study of magnetic interactions because of the very narrow fluorescence linewidth reported for it. A paper characterizing the interaction of magnetic fields with this laser material was subsequently published by Kiss*, including results of both homogeneous and inhomogeneous magnetic field modulation experiments.

The development of rare earth doped yttrium aluminum garnet (YAG) lasers by Bell Telephone Laboratories has placed a class of optically isotropic materials with reasonably narrow emission linewidths at our disposal. In particular $YAG:Nd^{3+}$ has been reported by Geusic et al.** to have relatively low threshold for CW operation even at room temperature, and sufficiently narrow emission lines at 77°K to make Zeeman effect measurements appear feasible.

Spectral data on such other new laser materials as LaF_3 doped with Nd^{3+} and Er^{3+} being developed at Varian Associates and Gd_2O_3 doped with Nd^{3+} and Eu^{3+} being developed at Korad are being sought in order to determine their possible suitability for magnetic modulation experiments.

B. Experimental Design for $YAG:Nd^{3+}$ Zeeman Study

In view of the lack of Zeeman data on $YAG:Nd^{3+}$, design of a system in which the Zeeman splitting of the laser transition can be measured has been undertaken.

Linde can supply continuous wave $YAG:Nd^{3+}$ laser rods, 3 cm long by 0.3 cm OD. Such a rod is now on order. In order to subject the rod to a homogeneous, high-intensity, magnetic field we have decided upon a Helmholtz coil configuration for producing a pulsed magnetic field. A sketch of the apparatus, which is nearing completion in the Research Department's Model Shop, is shown in Fig. 3. The design is similar to our inhomogeneous field apparatus, except that the larger coil diameter permits the pump lamp to be mounted inside the coils, thereby reducing shadowing. Calculations indicate that peak fields of 50 kgauss should readily be attainable with this apparatus.

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*Z. Kiss, Appl. Phys. Letters, 3, 145 (1963).

**J. E. Geusic, H. M. Marcos, and L. G. Van Uitert, Appl. Phys. Letters, 4, 182 (1964).

We were unable to find tabulations of the axial field intensity within Helmholtz coils. Therefore, the solution of the magnetic field equation:

$$B_x = \frac{\mu_0 I a^2}{2} \left\{ (a^2 + x^2)^{-3/2} + [(a^2 + (a - x)^2]^{-3/2} \right.$$
$$+ 3 r^2(a^2 - 4x^2)/4(a^2 + x^2)^{7/2}$$
$$\left. + 3 r^2(a^2 + 4(a - x)^2)/4(a^2 + (a - x)^2)^{7/2} \right\}$$

was programmed for solution by our IBM 1620 computer. In the equation, a is the coil radius, x is the perpendicular distance from the plane of symmetry between the two coils, and r is the radial distance from the axis through the centers of the coils. A plot of the values $2B_x/\mu_0 I$ for a = 1 is shown in Fig. 4. This indicates that even if the sample extends from the center of one coil to the center of the other, the maximum variation in magnetic field within the sample is only about 5%, which would not seriously broaden Zeeman components under conditions anticipated with YAG:Nd^{3+}.

It is planned to observe the Zeeman splitting by using the laser in the pulse pumped mode. The emission will pass through a Fabry-Perot etalon for spectral resolution and be observed during the magnetic field pulse by the STL streak camera. The S-1 image converter tube should have adequate response at 1.06 μ to permit such observations. Dielectric coated etalon reflectors with high reflectivity at 1.06 μ have been ordered from Perkin Elmer Corporation for this experiment. It is doubtful whether the fluorescent intensity in the absence of laser oscillation will be high enough to permit observation of the normal Zeeman splitting with the pulsed magnetic field.

If the pulsed experiments yield encouraging results, design of a CW magnetic modulated laser using YAG:Nd^{3+} will be undertaken, as specified in Phase II of the contract.

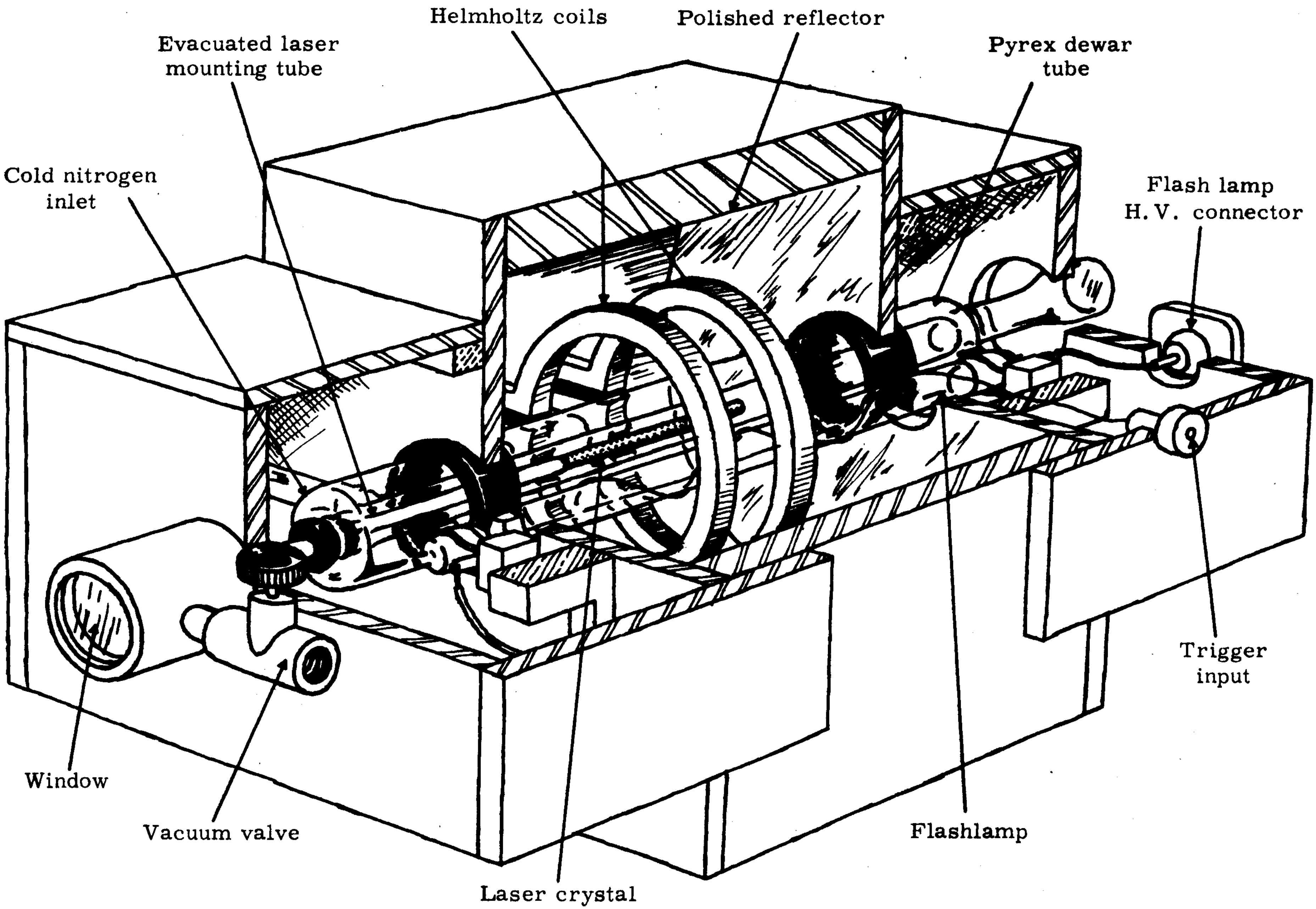

Fig. 3. Homogeneous magnetic field laser configuration.

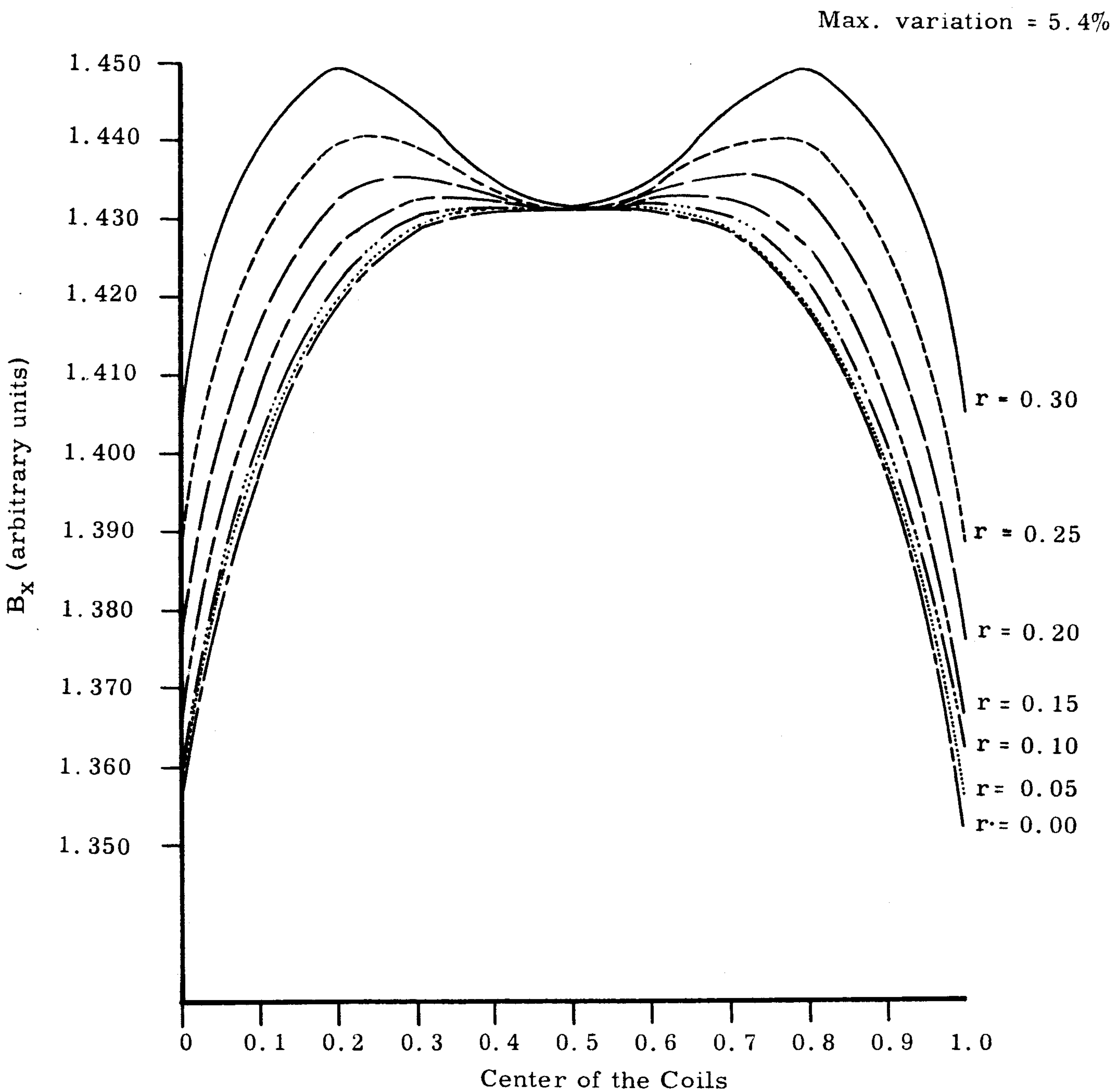

Fig. 4. Variation of the axial magnetic field within Helmholtz coils.